

(Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂: A Bulk Form Diluted Magnetic Semiconductor isostructural to the "32522" Fe-based Superconductors

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Abstract. - A new diluted magnetic semiconductor system, (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂, has been synthesized and characterized. 10% Mn substitution for Zn in bulk form (Sr₃La₂O₅)Zn₂As₂ results in a ferromagnetic ordering below Curie temperature, $T_C \sim 40$ K. (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂ has a layered crystal structure identical to that of 32522-type Fe-based superconductors, and represents the fifth DMS family that has a direct counterpart among the FeAs high temperature superconductor families.

Introduction. – The research of DMS (diluted magnetic semiconductors) has been explosive following the successful fabrication of III-V (Ga,Mn)As ferromagnetic thin films by Ohno et al in 1990's [1]. Over the past two decades, much progress has been made in the fabrication of DMS materials and the understanding of the ferromagnetism [2–6]. On the other hand, most extensively studied DMS materials are thin films that are grown under non-equilibrium condition, which encounters some inherent difficulties. For example in (Ga,Mn)As, some Mn impurities enter the interstitial sites, and makes it difficult to precisely determine the amount of Mn that substitutes ionic Ga, which donates a hole and acts as a local moment [6]. The thin films also prohibit the utilization of powerful magnetic probes such as neutron scattering and nuclear magnetic resonance (NMR) that are based on bulk form specimens, to provide complementary information for understanding the ferromagnetism at a microscopic level. Seeking for bulk form DMS system grown in thermally equilibrium condition will be helpful to understand the ferromagnetism.

Recently, through doping Mn into the I-II-V semiconductors LiZnAs and LiZnP, Deng et al successfully synthesized two bulk DMS systems, Li(Zn,Mn)As [7] and

Li(Zn,Mn)P [8], with $T_C \sim 50$ K. The I-II-V DMSs have advantages of decoupling spins and carriers, where spins are introduced by Mn atoms and carriers are created by off-stoichiometry of Li concentrations. This advantage makes it possible to precisely control the amount of spins and carriers, and investigate their individual effects on the ferromagnetic ordering. More recently, several more bulk DMS systems have been reported. Firstly, Ding et al reported the ferromagnetic ordering below $T_C \sim 40$ K in a "1111" type (La,Ba)(Zn,Mn)AsO system [9]; Han et al reported the ferromagnetism in (La,Ca)(Zn,Mn)SbO semiconductor [10] and Yang et al reported the fabrication of (La,Sr)(Cu,Mn)SO DMS with $T_C \sim 210$ K [11]. Secondly, Zhao et al. reported the "122" type DMS systems, (Ba,K)(Zn,Mn)₂As₂, which has T_C as high as 180 K [12], and Yang et al observed the ferromagnetic transition below $T_C \sim 17$ K and a large negative magnetoresistance in (Ba,K)(Cd,Mn)₂As₂ [13]. The Curie temperature of (La,Sr)(Cu,Mn)SO and (Ba,K)(Zn,Mn)₂As₂ polycrystals is already comparable to the record T_C of (Ga,Mn)As thin films [14, 15].

The availability of bulk form DMS specimens readily enables the microscopic investigation by μ SR (muon spin relaxation) and NMR techniques. μ SR has demonstrated that the exchange interaction supporting ferromagnetic coupling in Li(Zn,Mn)As, (La,Ba)(Zn,Mn)AsO,

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(Ba,K)(Zn,Mn)₂As₂ and (Ga,Mn)As has a common origin and comparable magnitude for a given spatial density of ordered moments, no matter the specimens are thin films or bulk forms [7, 9, 12, 16]. Moreover, Ding *et al.* has conducted ⁷Li NMR measurement of Li(Zn,Mn)P, and successfully detected the fast relaxed Li sites that have Mn at nearest neighbor sites. They found that the Mn spin-spin interactions extend over many unit cells, which explains why DMSs could exhibit a relatively high T_C with such a low density of Mn [17].

More interestingly, each of above “111”, “1111” and “122” DMSs families has a direct counterpart among the FeAs-based high temperature superconductor families. For example, “1111”-type (La,Ba)(Zn,Mn)AsO DMS has a ZrCuSiAs-type tetragonal structure, identical to FeAs-based “1111” type LaFeAsO_{1-x}F_x high temperature superconductor ($T_c = 26$ K) [18] and the antiferromagnetic LaMnAsO ($T_N = 317$ K) [19]. The excellent lattice matching (lattice constants are within 5% difference) between ferromagnetic, antiferromagnetic and superconducting systems opens the possibilities to make junctions between these systems through the As layer. The parameters for all these compounds are listed in Table. 1. We also mention that for the “11” type Fe-based superconductor FeSe [20], the counterpart is (Zn,Mn)Se, which has been extensively studied as one of the prototypical II-VI DMSs.

In this letter, we report successful synthesis and characterization of a new bulk DMS system, (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂, which is isostructural to the “32522” FeAs-based superconductor (Ca₃Al₂O₅)Fe₂As₂ [27]. 5% Mn substitution for Zn in the parent semiconductor (Sr₃La₂O₅)Zn₂As₂ results in ferromagnetic ordering, as indicated by the strong enhancement of magnetization below $T_C \sim 36$ K. The bifurcation of ZFC and FC curves below $T_f = 12.5$ K and a parallelogram-shaped hysteresis loop are also observed. T_C increases to 40 K with the doping level increasing to 10%, but starts to decrease at the doping level of 20%. The saturation moment is suppressed from 0.5 μ_B /Mn for $x = 0.10$ to 0.2 μ_B /Mn for $x = 0.20$. More Mn doping also suppresses the coercive field from 324 mT for $x = 0.05$ to 111 mT for $x = 0.20$. No ferromagnetic ordering is observed for the doping level of 30% Mn.

Experimental methods. – We synthesized (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂ ($x = 0.00, 0.05, 0.10, 0.20, 0.30$) polycrystalline specimens by the solid state reaction method. High purity elements of La, Zn, Mn and As were mixed and heated to 900 °C in an evacuated silica tube to produce intermediate products LaAs, ZnAs and MnAs. They were then mixed with La₂O₃, SrO with nominal concentrations and slowly heated up to 1150 °C, and held for 50 hours before cooling to room temperature with turning off the furnace. The polycrystals were characterized by X-ray diffraction at room temperature and dc magnetization by Quantum Design SQUID. The electrical resistance was measured on sintered pellets with

typical four-probe method.

Results and discussion. – We show the crystal structure of (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂ and the X-ray diffraction patterns in Fig. 1. Appreciable Bragg peaks from the parent compound (Sr₃La₂O₅)(Zn₂As₂) can be indexed by a layered tetragonal crystal structure (I_4/mmm), with $a = 4.2612$ Å and $c = 27.675$ Å. These lattice constants are close to $a = 4.069$ Å and $c = 26.876$ Å of a “32522” compound (Sr₃Sc₂O₅)(Fe₂As₂) [29], as well as $a = 3.742$ Å and $c = 26.078$ Å of the superconducting sample (Ca₃Al₂O₅)Fe₂As₂ [27]. We observed secondary phases of Zn₃As₂ and Sr₂As for $x \leq 0.10$, as marked by the stars and the arrows in Fig.1. These impurities are non-magnetic, which will not affect the magnetic properties discussed in the following section.

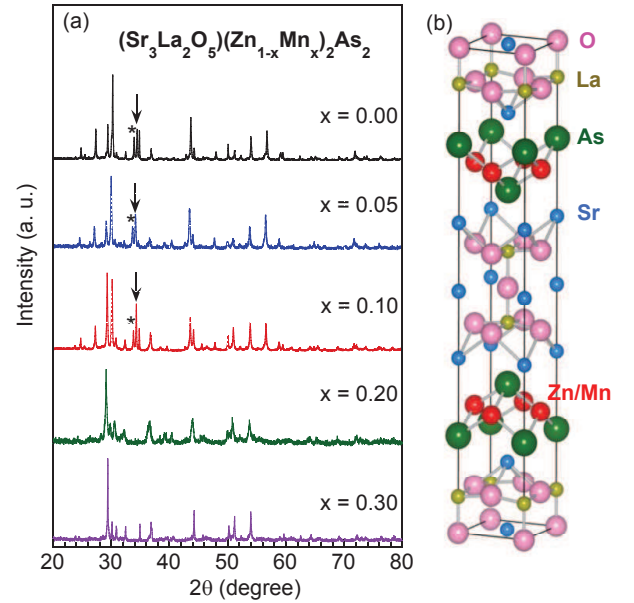


Fig. 1: (Color online) X-ray diffraction pattern (a) and crystal structure (b) of (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂. Secondary phases of Sr₂As (↓) and Zn₃As₂ (*) are marked for $x \leq 0.10$.

In Fig. 2, we show the electrical resistivity measured for (Sr₃La₂O₅)(Zn_{1-x}Mn_x)₂As₂ with $x = 0.05, 0.10, 0.20, 0.30$. The resistivity of all samples monotonically increases toward lower temperature. This type of behavior in Mn doped specimens has been observed in heavily doped region of (Ga_{1-x}Mn_x)As [6], as well as (La,Ba)(Zn,Mn)AsO [9] and (Ba,K)(Zn,Mn)₂As₂ [12] DMS polycrystals. It has been ascribed to the scattering of carriers by the magnetic fluctuations through exchange interactions in (Ga_{1-x}Mn_x)As [6]. We have also conducted the Hall effect measurements for the sample of (Sr₃La₂O₅)(Zn_{0.90}Mn_{0.10})₂As₂. The large resistivity forbids us to accurately determine the carriers type, and a preliminary measurement indicates that the carrier den-

--	SC	DMS	AFM
"11"	FeSe ($T_c \sim 8\text{K}$) [20] Tetragonal $a=3.7676\text{\AA}$ $c=5.4847\text{\AA}$	(Zn,Mn)Se ($T_f \sim 24\text{K}$) [21] Cubic $a=5.669\text{\AA}$	MnSe ($T_N \sim 197\text{K}$) [22] Cubic $a=5.464\text{\AA}$
"111"	LiFeAs ($T_c \sim 18\text{K}$) [23] Tetragonal $a=3.77\text{\AA}$ $c=6.36\text{\AA}$	Li(Zn,Mn)As ($T_C \sim 50\text{K}$) [7] Cubic $a=5.94\text{\AA}$	LiMnAs ($T_N \sim 393\text{K}$) [24] Tetragonal $a=4.273\text{\AA}$ $c=12.370\text{\AA}$
"1111"	LaFeAs(O,F) ($T_c \sim 26\text{K}$) [18] Tetragonal $a=4.0320\text{\AA}$ $c=8.7263\text{\AA}$	(La,Ba)(Zn,Mn)AsO ($T_C \sim 40\text{K}$) [9] Tetragonal $a=4.116\text{\AA}$ $c=9.11\text{\AA}$	LaMnAsO ($T_N \sim 317\text{K}$) [19] Tetragonal $a=4.11398\text{\AA}$ $c=9.03044\text{\AA}$
"122"	(Ba,K)Fe ₂ As ₂ ($T_c \sim 38\text{K}$) [25] Tetragonal $a=3.917\text{\AA}$ $c=13.2968\text{\AA}$	(Ba,K)(Zn,Mn) ₂ As ₂ ($T_C \sim 180\text{K}$) [12] Tetragonal $a=4.131\text{\AA}$ $c=13.481\text{\AA}$	BaMn ₂ As ₂ ($T_N \sim 625\text{K}$) [26] Tetragonal $a=4.1684\text{\AA}$ $c=13.4681\text{\AA}$
"32522"	Ca ₃ Al ₂ O _{5-y} Fe ₂ As ₂ ($T_c \sim 30.2\text{K}$) [27] Tetragonal $a=3.742\text{\AA}$ $c=26.078\text{\AA}$	Sr ₃ La ₂ O ₅ (Zn,Mn) ₂ As ₂ ($T_C \sim 40\text{K}$, this work) Tetragonal $a=4.2612\text{\AA}$ $c=27.675\text{\AA}$	hypothetical (Sr ₃ La ₂ O ₅ Mn ₂ As ₂)
"42622"	Sr ₄ V ₂ O ₆ Fe ₂ As ₂ ($T_c \sim 37.2\text{K}$) [28] Tetragonal $a=3.9296\text{\AA}$ $c=15.6732\text{\AA}$	Sr ₄ Ti ₂ O ₆ (Zn,Mn) ₂ As ₂ ($T_C \sim 25\text{K}$) (Unpublished)	hypothetical (Sr ₄ Ti ₂ O ₆ Mn ₂ As ₂)

Table 1: The transition temperature T_c for a superconductor (SC), Curie temperature T_C for a diluted magnetic semiconductor (DMS) and Neel temperature T_N for an antiferromagnet (AFM) of "11", "111", "1111", "122", "32522" and "42622" type compounds. The type of crystal structure and lattice constants are also listed for available compounds.

sity is in the order of 10^{16} cm^{-3} . This carrier density is comparable to that of $\text{Li}_{1.1}\text{Zn}_{1-x}\text{Mn}_x\text{P}$ [8] but 4 orders smaller than that of $\text{Li}_{1.1}\text{Zn}_{1-x}\text{Mn}_x\text{As}$ [7].

In Fig. 3, we show the zero-field cooled (ZFC) and field cooled (FC) measurements of the dc -magnetization M of $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ for $B_{ext} = 0.1$ Tesla. For the doping of $x = 0.05$, we observe a strong increase of M at $T_C = 36$ K, and the bifurcation of ZFC and FC curves below the temperature $T_f = 12.5$ K, where T_f stands for the freezing temperature of individual spins or domain wall motion. The saturation moment at 2 K is $0.4 \mu_B/\text{Mn}$. With the doping level increasing to $x = 0.10$, T_C increases to ~ 40 K, and the saturation moment increases to $0.5 \mu_B/\text{Mn}$. This indicates that additional Mn atoms raise the ferromagnetic ordering temperature. Both T_C and moment size start to decrease with further doping to $x = 0.20$. The ferromagnetic ordering disappears for the doping level of $x = 0.30$. This is primarily due to the competition of antiferromagnetic exchange interaction between spins from

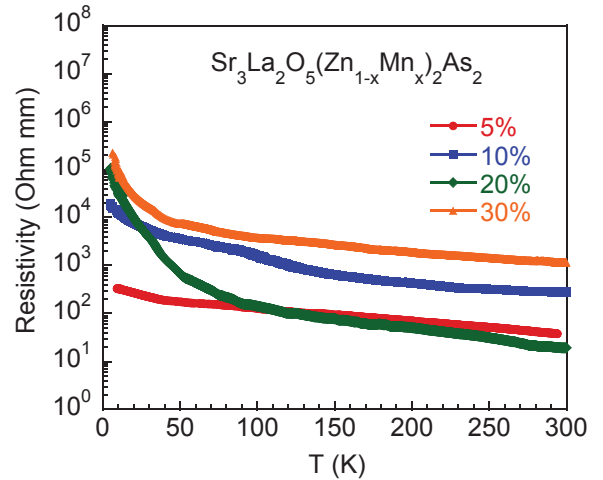


Fig. 2: (Color online) The electrical resistivity of $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ with $x = 0.05, 0.10, 0.20, 0.30$.

nearest neighbor Mn sites. For 100% Mn substitution for Zn in "11", "111", "1111" and "122" compounds, the ending product is always an antiferromagnet with Neel temperature $T_N \sim 200$ K - 600 K, as displayed in Table. 1. Following this trend, we would expect a hypothetical antiferromagnet with "32522" structure, $(\text{Sr}_3\text{La}_2\text{O}_5)\text{Mn}_2\text{As}_2$. We fit the temperature dependence of M above T_C to a Curie-Weiss law. The effective paramagnetic moment is determined to be $5 \sim 6\mu_B/\text{Mn}$, indicating that the valence of Mn ions are +2, and it is in a high spin state, as observed in other Mn doped DMSs [5–7].

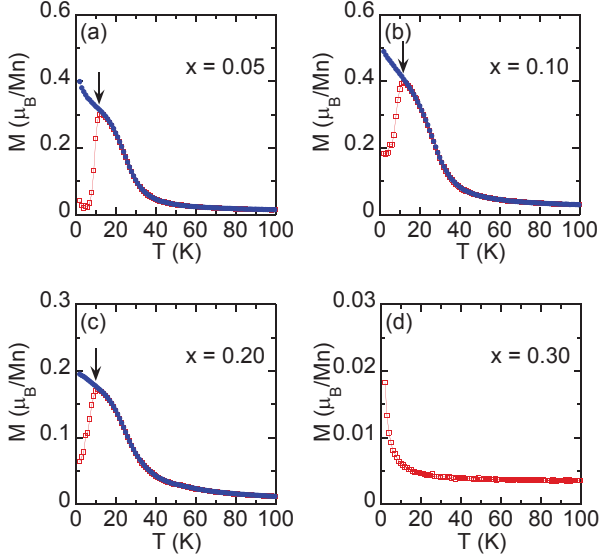


Fig. 3: (Color online) The magnetization M for $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ with $x = 0.05, 0.10, 0.20, 0.30$ obtained in the zero field cooling (ZFC, in red) and field cooling (FC, in blue) mode under the external field of 0.1 Tesla. Arrows mark the position of T_f . We note that T_C is defined as the temperature where M shows a sharp upturn in the low field of 1 mT (not shown), higher magnetic field suppresses the sharp upturn feature.

In Fig. 4, we show the isothermal magnetization of $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$. For $x = 0.05$, a parallelogram-shaped hysteresis loop with a coercive field of 324 mT is observed at 5 K. The coercive field continuously decreases to 111 mT with the doping level increasing to $x = 0.20$. The coercive fields are larger than $\sim 5 - 10$ mT of the cubic structural $(\text{Ga}_{0.965}\text{Mn}_{0.035})\text{As}$ [1], $\text{Li}_{1.1}(\text{Zn}_{0.97}\text{Mn}_{0.03})\text{As}$ [7], and $\text{Li}_{1.1}(\text{Zn}_{0.97}\text{Mn}_{0.03})\text{P}$ [8], but smaller than ~ 1000 mT of the two dimensional $(\text{La},\text{Ba})(\text{Zn},\text{Mn})\text{AsO}$ [9] and $(\text{Ba},\text{K})(\text{Zn},\text{Mn})_2\text{As}_2$ [12]. We show the temperature dependence of the hysteresis loop for $x = 0.10$ in Fig. 4(d). The coercive field decreases from 203 mT to 42 mT at 10 K and becomes zero at 60 K.

As we have discussed in ref. [9], the bifurcation of ZFC and FC curves and the hysteresis loops can be found not only in regular ferromagnets [30] but also in spin glasses [31]. Neutron scattering technique can

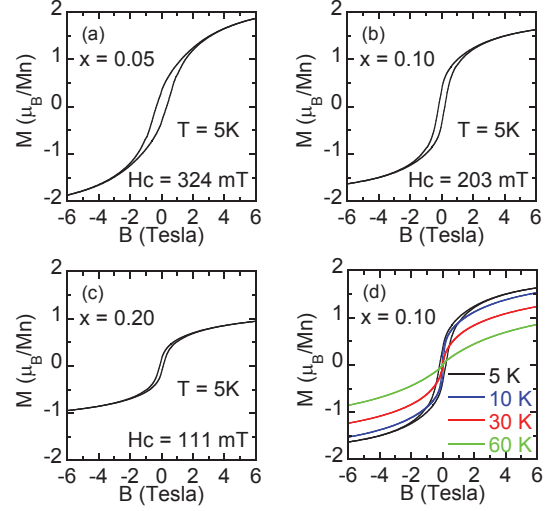


Fig. 4: (Color online) The isothermal magnetization for $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ with $x = 0.05, 0.10, 0.20$ measured at 5 K, and for $x = 0.10$ measured at 5 K, 10 K, 30 K and 60 K. H_c values are the coercive field at 5 K.

resolve spatial spin correlations and decisively distinguish the two cases. Our neutron diffraction experiments on polycrystals $(\text{Ba}_{0.7}\text{K}_{0.3})(\text{Zn}_{0.9}\text{Mn}_{0.1})_2\text{As}_2$ [12] and $(\text{La}_{0.9}\text{Sr}_{0.1})(\text{Zn}_{0.9}\text{Mn}_{0.1})\text{AsO}$ [32] were not able to decouple the magnetic and structural Bragg peaks since they superpose to each other. Single crystals are expected for high resolution neutron scattering experiments. None the less, the moment size is usually small for typical spin glasses, i.e., $\sim 0.01 \mu_B/\text{Mn}$ for the II-VI $(\text{Zn},\text{Mn})\text{Se}$ or other typical dilute alloy spin glasses [33–35]. Considering that the saturation moment size of our $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ is as large as $0.5 \mu_B$, we tentatively assign it to a ferromagnetic ordering rather than a spin glass.

The ferromagnetism in various diluted magnetic semiconductors and oxides has been explained by several theoretical models, such as Zener’s model [36], percolation of bound magnetic polarons (BMPs) [37–39], and $d-d$ double exchange due to hopping between transition metal d states [40]. In the case of $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$, the resistivity is large ($\sim 10^4 \Omega \text{ mm}$ at 2 K) and the carrier density is $\sim 10^{16} \text{ cm}^{-3}$. The low carrier density and the relative low T_C seems more amenable with the BMPs model.

Summary. — In summary, we report the synthesis and characterization of bulk form diluted magnetic semiconductor $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ with the Curie temperature ~ 40 K. The physical properties of this layered polycrystal are very similar to the recently discovered bulk form DMSs, $(\text{La},\text{Ba})(\text{Zn},\text{Mn})\text{AsO}$ [9] and $(\text{Ba},\text{K})(\text{Zn},\text{Mn})_2\text{As}_2$ [12]. Summing up previously reported “11” type $(\text{Zn},\text{Mn})\text{Se}$ (II-VI), “111” type $\text{Li}(\text{Zn},\text{Mn})\text{As}$, “1111” type $(\text{La},\text{Ba})(\text{Zn},\text{Mn})\text{AsO}$

and "122" type $(\text{Ba,K})(\text{Zn,Mn})_2\text{As}_2$, the "32522" type $(\text{Sr}_3\text{La}_2\text{O}_5)(\text{Zn}_{1-x}\text{Mn}_x)_2\text{As}_2$ system in current study represents the fifth DMS system that has a direct counterpart with identical/similar structure in the Fe-based superconductors. As we stated in earlier papers [7, 9, 12], the common structure and excellent lattice matching between ferromagnetic, antiferromagnetic and superconducting systems make it possible to make junctions between these systems through the As layer.

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